

Possible Pairing Symmetry of Superconductor $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ Yunori NISIKAWA^{*1}, Hiroaki IKEDA² and Kosaku YAMADA²¹*Synchrotron Radiation Research Center, Japan Atomic Energy Research Institute, Mikazuki, Sayo, Hyogo 679-5148*²*Department of Physics, Kyoto University, Kyoto 606-8502*

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To discuss the possibility that the superconductivities in $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ are induced by electron correlation, we investigate the possible pairing symmetry based on the single-band Hubbard model whose dispersion of the bare energy band is obtained using the band structure calculation of $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$. The superconducting transition temperature is estimated by solving the Éliashberg equation. In this equation, both normal and anomalous self-energies are calculated up to the third-order terms with respect to the Coulomb repulsion. In the case of spin-singlet pairing, the candidate of pairing symmetry (the maximum eigen value $\lambda_{\text{max}}^{\text{SS}}$ of Éliashberg's equation) belongs to the d -wave(E_2 representation of D_6 group). In the case of spin-triplet pairing, the candidate of pairing symmetry (the maximum eigen value $\lambda_{\text{max}}^{\text{ST}}$ of Éliashberg's equation) belongs to the $f_{y(y^2-3x^2)}$ -wave (B_1 representation of D_6 group). It is found that $\lambda_{\text{max}}^{\text{SS}} \simeq \lambda_{\text{max}}^{\text{ST}}$ and the transition temperatures of unconventional pairing state are estimated to be low compared with the observed temperature within our simple model.

KEYWORDS: $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$, superconductivity, vertex correction

Recently, Tanaka *et al.* discovered superconductivity in $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ ($x \simeq 0.35, y \simeq 1.4$) with the transition temperature $T_c \simeq 5$ K.¹ $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ consists of a two-dimensional triangular lattice of cobalt ions formed by a network of edge-sharing CoO_6 octahedra, separated by layers of Na ions and H_2O molecules. The superconductivity in this compound has been intensively investigated.²⁻¹⁹ The normalized electronic specific heat data in the superconducting state well fit the T^3 dependence.¹² The NMR/NQR measurements have been performed by several groups.¹³⁻¹⁵ The conclusions of the groups have been contradictory regarding the pairing symmetry of the superconductivity, to date. Fujimoto *et al.* observed an absence of the coherence peak just below T_c ¹⁵ and quite recently, their observation has been confirmed by Ishida *et al.*¹⁶ These results may suggest the possibility of unconventional superconductivity in $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$. In this paper, we discuss the possibility that the superconductivities in $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ are induced by electron correlation. In our previous study,¹⁷ we investigated the superconducting instabilities of a two-dimensional triangular lattice in the Hubbard model with the next nearest hopping integral, by the perturbation theory with respect to

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Coulomb repulsion U and fluctuation-exchange approximation(FLEX). We have found that f -wave spin-triplet pairing is the most stable in wide ranges of next nearest neighbor hopping integrals and electron number densities. The perturbation approach is sensitive to the dispersion of the bare energy band, by its nature. This implies that the lattice structures and the band filling play essential roles in the calculation of T_c , and it is important to evaluate T_c on the basis of the detailed electronic structure in each system. Therefore, we construct a two-dimensional Hubbard model whose dispersion of the bare energy band is obtained using the band structure calculation of $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$. Based on this Hubbard model, we estimate T_c by the perturbation theory with respect to Coulomb repulsion U .

Here, we briefly mention the band structure calculation of $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ performed by Nisikawa *et al.*²⁰ They used the full-potential linearized augmented plane wave (FLAPW) method in the local density approximation(LDA). In their calculation, the H_2O molecules were treated as mere spacers which realistically separate two CoO_2 layers in a hydrated compound and the Na ions were treated using the virtual crystal approximation. The obtained band structure has two-dimensional properties. The Fermi surfaces consist of a large “troche”-like hole Fermi surface around the Γ point and small cylindrical hole Fermi surfaces near the K points. The Fermi level is very close to the van Hove singularity. Co t_{2g} orbitals take the main part of the density of states near the Fermi energy. The obtained Fermi surface is similar to the Fermi surface of nonhydrated $\text{Na}_{0.5}\text{CoO}_2$ obtained by Singh.²¹ But the volume of our cylindrical hole Fermi surface near the K point is larger than the volume of Singh’s one. There is no experimental measurements about Fermi surface of $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ to date. Fermi surface is one of the important matters to consider in investigating the mechanism of superconductivity. So comparison of band structure calculation and experiments is desired.

We start from quasi-particle state and use the single-band Hubbard model for discussing the mechanism of superconductivity. We obtain the dispersion $\epsilon(\mathbf{k})$ of the bare energy band by using the band structure calculation of $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ ($x = 0.35$). $\epsilon(\mathbf{k})$ can be expanded as $\epsilon(\mathbf{k}) = -\sum_{\mathbf{R} \in L} t_{\mathbf{R}} \exp(i\mathbf{R} \cdot \mathbf{k})$, where L is the set of lattice vectors of a two-dimensional triangular lattice. We rescale length, energy, temperature and time by $a, t, \frac{t}{k_B}, \frac{\hbar}{t}$ respectively (where a, t, k_B, \hbar are the lattice constants of the hexagonal Co plane, third hopping integral, Boltzmann constant and Planck constant divided by 2π , respectively). Our Hamiltonian is written as

$$H = \sum_{\mathbf{k}, \sigma} (\epsilon(\mathbf{k}) - \mu) a_{\mathbf{k}\sigma}^\dagger a_{\mathbf{k}\sigma} + \frac{U}{2N} \sum_{\sigma \neq \sigma'} \sum_{\mathbf{k}_1 \mathbf{k}_2 \mathbf{k}_3 \mathbf{k}_4} \delta_{\mathbf{k}_1 + \mathbf{k}_2, \mathbf{k}_3 + \mathbf{k}_4} a_{\mathbf{k}_1 \sigma}^\dagger a_{\mathbf{k}_2 \sigma'}^\dagger a_{\mathbf{k}_3 \sigma'} a_{\mathbf{k}_4 \sigma},$$

where μ and U are the chemical potential and the Coulomb repulsion, respectively. The coefficients $t_{\mathbf{R}}$ are shown in Fig. 1. We calculate T_c by solving Éliashberg’s equation (Fig. 2(a)). In the equation, the normal self-energy and the effective interaction are obtained within the third-order perturbation with respect to U (Figs. 2(b) and 2(c)). The diagrams enclosed by

a dashed line in Fig. 2(c) are the vertex correction terms which are not direct contributions from spin fluctuations. The other diagrams are included in RPA and FLEX. We call the latter 'RPA-like diagrams' in this paper. To satisfy Luttinger's theorem, that is, the conservation law of particle number, we adjust the chemical potential μ using the secant method. To solve Éliashberg's equation by using the power method algorithm, we have to calculate the summation over the momentum and the frequency space. Since all summations are in convolution forms, we can carry them out using the algorithm of the fast Fourier transformation. For the frequency, irrespective of the temperature, we have 1024 Matsubara frequencies. Therefore, we calculate throughout in the temperature region $T \geq T_{\text{lim}}$, where T_{lim} is the lower limit temperature for reliable numerical calculation, which is estimated to be approximately $2.5 \times 10^{-3} (> W/(2\pi \times 1024) \simeq 2.2 \times 10^{-3})$, where W is the bandwidth; we divide a primitive cell into 128×128 meshes.

The possible five candidates of unconventional pairing symmetry in our two-dimensional model are shown in Fig. 3. In our calculation, we obtain the maximum eigen value $\lambda_{\text{max}}^{\text{SS}}$ of spin-singlet pairing belonging to the d -wave and the maximum eigen value $\lambda_{\text{max}}^{\text{ST}}$ of spin-triplet pairing belonging to the $f_{y(y^2-3x^2)}$ -wave. The $f_{y(y^2-3x^2)}$ -wave pairing state has been proposed in several studies of other similar models.^{17,18,22} The calculated eigenvalues $\lambda_{\text{max}}^{\text{ST}}$ and $\lambda_{\text{max}}^{\text{SS}}$ for various values of T are shown in Fig. 4(a). From Fig. 4(a), we can see that when we estimate $W (\simeq 13t_3) \simeq 0.5$ eV from band structure calculation, our T_c is estimated to be far below 1 K, which is low compared with the observed $T_c \simeq 5$ K.

To examine how the vertex corrections influence T_c , we calculate the eigenvalues $\lambda_{\text{RPA-like}}$ of Éliashberg's equation by including only RPA-like diagrams of anomalous self-energies up to the third order. We compare $\lambda_{\text{RPA-like}}$ with λ_{TOPT} calculated by including full diagrams of anomalous self-energies up to third order. The calculated eigenvalues $\lambda_{\text{RPA-like}}^f$ (for f -wave triplet pairing) and $\lambda_{\text{RPA-like}}^d$ (for d -wave singlet pairing) are shown in Fig. 4(b). In RPA-like calculation, the momentum dependence of static bare susceptibility $\chi_0(\mathbf{q}, 0)$ is important for pairing symmetry. The calculated results of $\chi_0(\mathbf{q}, 0)$ are shown in Fig. 5. In this figure, we can observe a broad peak around M the points and a sharp peak around the Γ point. From Fig. 4(b), we can see that the vertex correction terms reduce the eigenvalues of spin-singlet pairing, on the other hand in the case of spin-triplet pairing, vertex correction terms work in favor of realizing the spin-triplet superconductivity. The fact that the vertex correction for a simple one-boson propagation is important for spin-triplet pairing, is the same as the case of Sr_2RuO_4 .²³ As a result, we have $\lambda_{\text{max}}^{\text{SS}} \simeq \lambda_{\text{max}}^{\text{ST}}$ within our simple model.

We also calculate the self-energy $\Sigma_n^R(\mathbf{k}, \omega)$ using the Padé approximation (whose results are not presented in this paper). The ω -dependence of both $\text{Re}\Sigma_n^R(\mathbf{k}_f, \omega)$ and $\text{Im}\Sigma_n^R(\mathbf{k}_f, \omega)$ (where \mathbf{k}_f is a Fermi momentum) near $\omega = 0$, are respectively given by $\text{Re}\Sigma_n^R(\mathbf{k}_f, \omega) \propto -\omega$ and $\text{Im}\Sigma_n^R(\mathbf{k}_f, \omega) \propto -\omega^2$. This behavior is the same as that for a conventional Fermi liquid.

Therefore, the perturbation treatment seems to work well in our calculation.

In summary, we have investigated the possibility of unconventional superconductivity originating from the electron correlation effects, on the basis of the Hubbard model whose dispersion of the bare energy band is obtained using the band structure calculation of $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$. In the case of spin-singlet pairing, we have obtained the maximum eigen value $\lambda_{\text{max}}^{\text{SS}}$ belonging to the d -wave. In the case of spin-triplet pairing, we have obtained the maximum eigen value $\lambda_{\text{max}}^{\text{ST}}$ belonging to the $f_{y(y^2-3x^2)}$ -wave. It is found that the vertex correction terms reduce eigenvalues of spin-singlet pairing, on the other hand in the case of spin-triplet pairing, vertex correction terms work in favor of realizing the spin-triplet superconductivity. As a result, it was found that $\lambda_{\text{max}}^{\text{SS}} \simeq \lambda_{\text{max}}^{\text{ST}}$ and the superconducting transition temperatures are estimated to be low compared with the observed $T_c \simeq 5\text{K}$ within our simple model. So we cannot decide spin-singlet pairing or spin-triplet pairing, and can not get reasonable transition temperatures of unconventional pairing state. This fact indicates that we have to use more a realistic model such as a multi-orbital model or the other mechanism such as the phonon-mediated mechanism may become dominant for the superconductivity in real system.¹⁴

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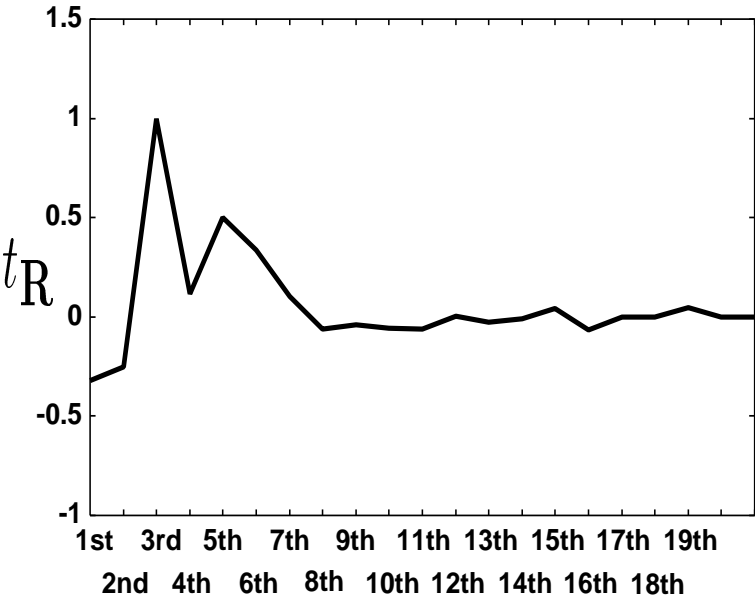


Fig. 1. The values of the coefficients t_R

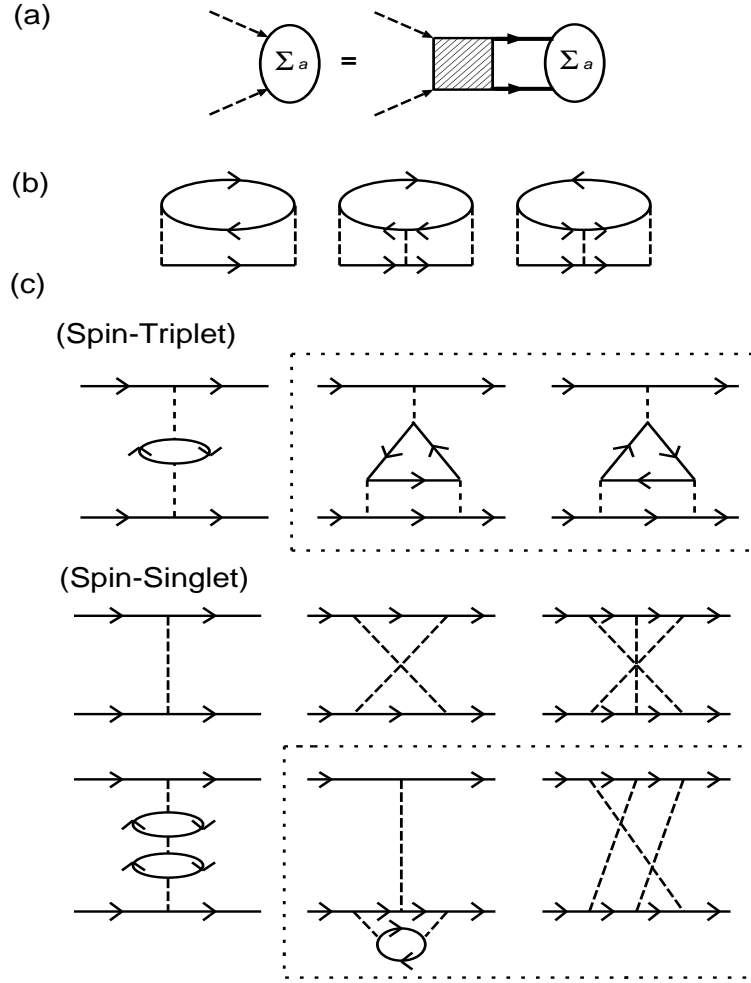


Fig. 2. (a)Éliashberg's equation. The thick line represents Green's function with self-energy correction. The shaded rectangle represents the effective interaction.(b) Feynman diagrams of the normal self-energy up to the third order. (c) Feynman diagrams of the effective interaction up to the third order. Solid and dashed lines correspond to the bare Green's function and the interaction, respectively. We omit writing the diagrams given by turning the vertex correction terms in this figure upside down.

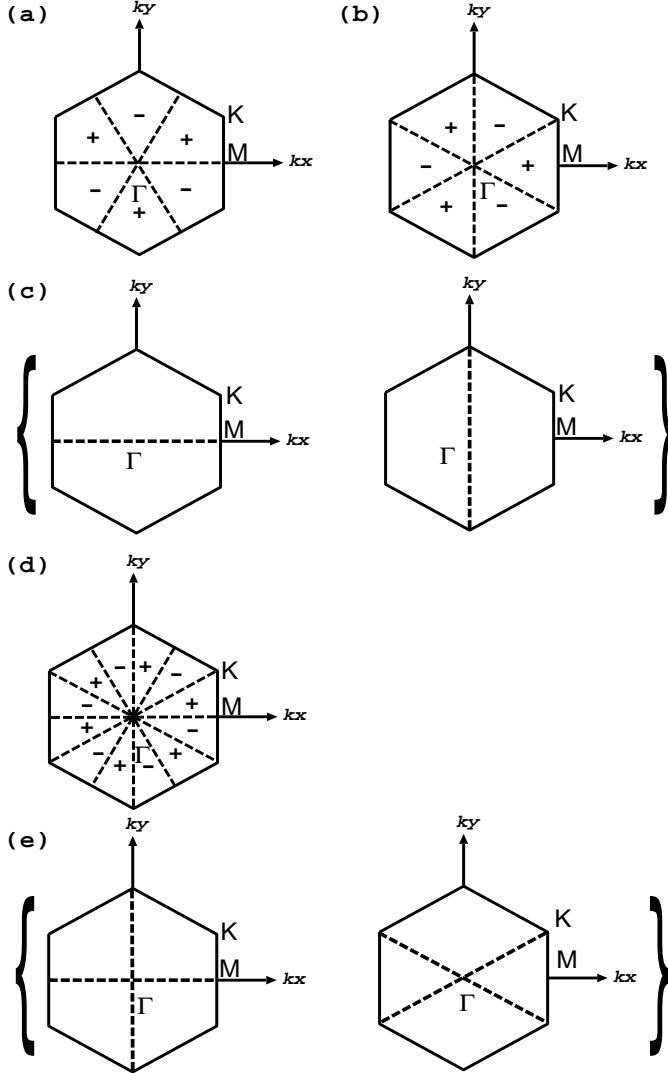


Fig. 3. The possible five candidates of unconventional pairing symmetry in our model.(a)spin-triplet $f_y(y^2-3x^2)$ -wave(B_1 representation of D_6 group), (b)spin-triplet $f_x(x^2-3y^2)$ -wave(B_2 representation of D_6 group), (c)spin-triplet p -wave(E_1 representation of D_6 group), (d)spin-singlet $i_{xy}(x^2-3y^2)(y^2-3x^2)$ -wave(A_2 representation of D_6 group), (e)spin-singlet d -wave(E_2 representation of D_6 group)

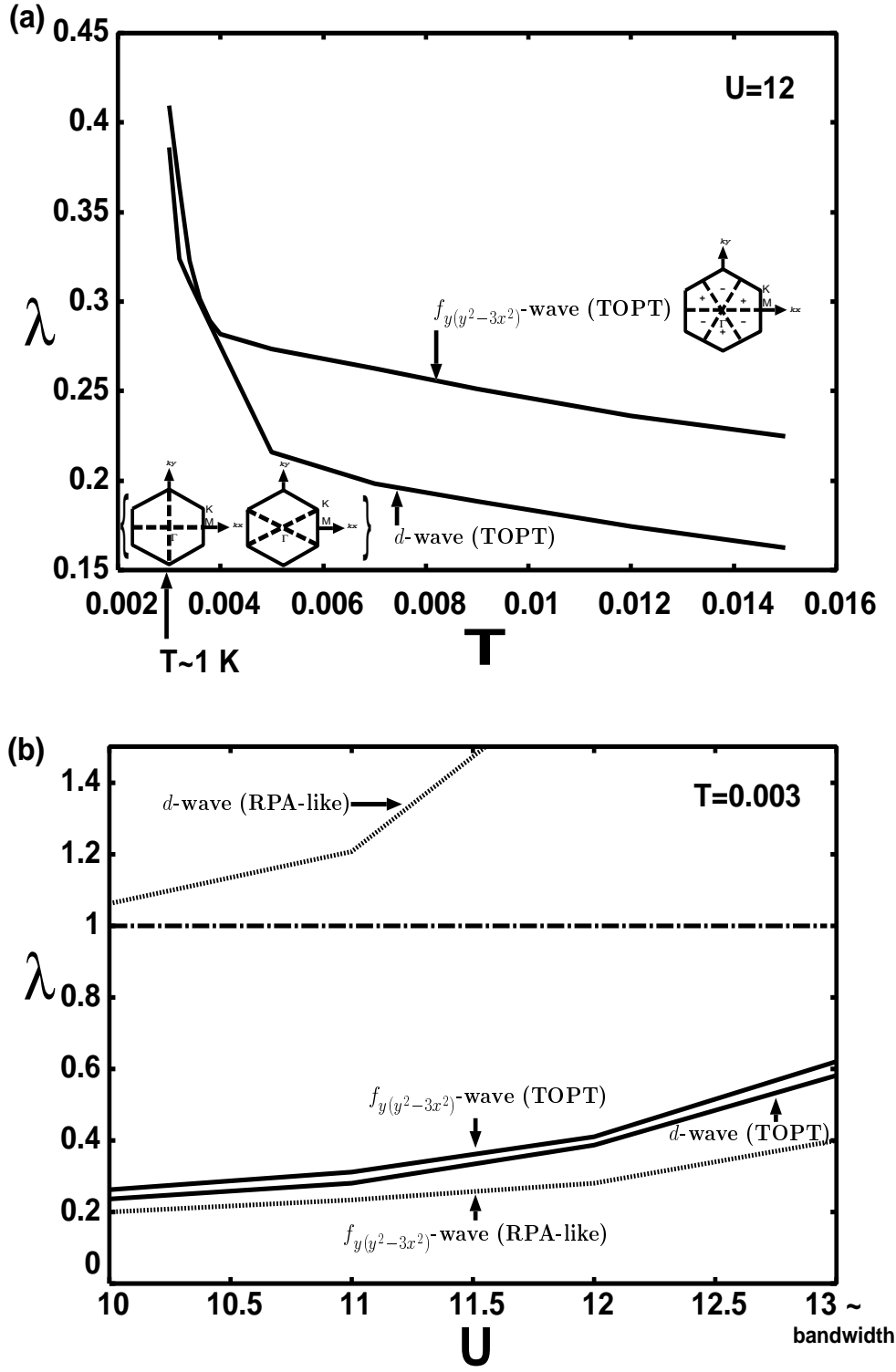


Fig. 4. (a) The calculated eigenvalues for various values of T (b) The calculated eigenvalues λ_{TOPT} and $\lambda_{\text{RPA-like}}$ for various values of U

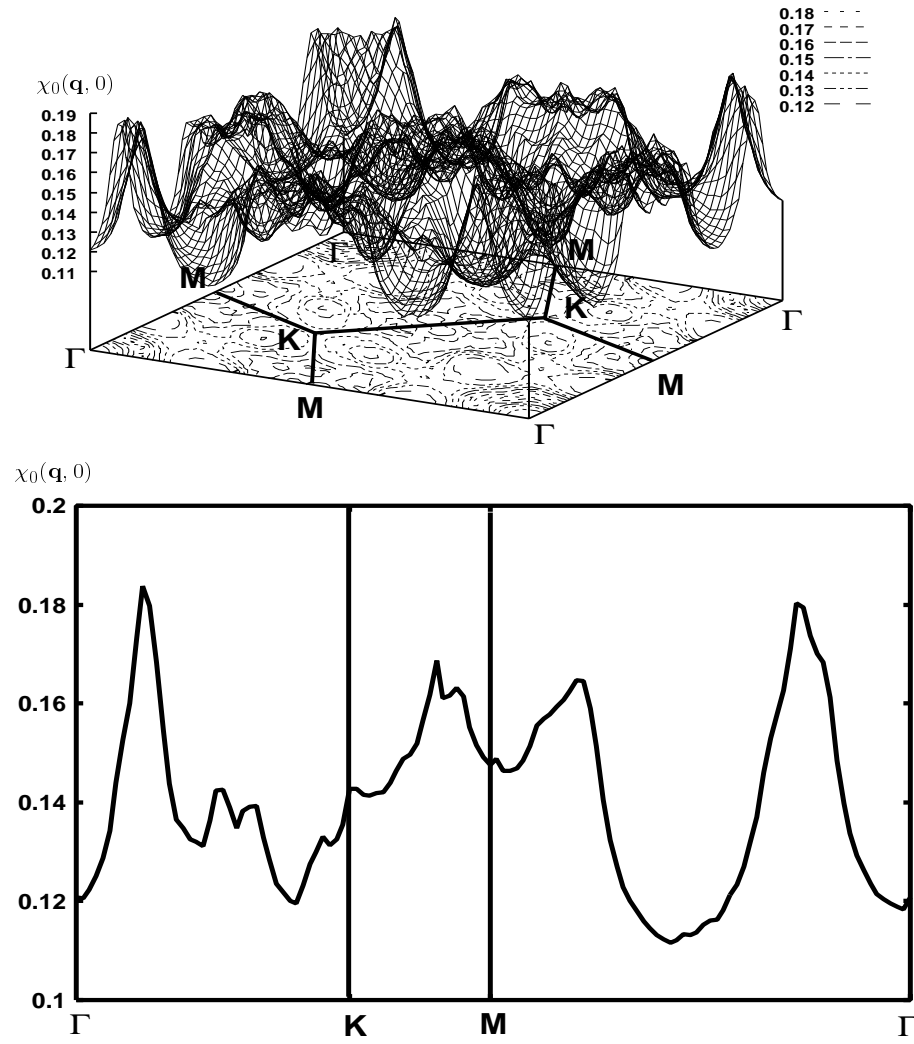


Fig. 5. The momentum dependence of the static bare susceptibility.